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## DETERMINATION OF THE OPTICAL CHARACTERISTICS OF COMPLEXLY STRUCTURED, LOCALLY MODIFIED REGIONS IN QUARTZOID GLASS

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The focal length and numerical aperture of a complexly structured modified region in a plate of porous glass are determined. It is shown that when a plate of porous glass with a modified region is baked the complex structure of the modified region remains. The focal length and numerical aperture of the complexly structure modified region in the quartzoid glass plate are determined. It is found that baking a porous glass plate with a modified region in a furnace considerably reduces the focal length and increases the numerical aperture to 0.9.

Key words: locally modified region, region of complex structure, quartzoid glass, optical characteristics.

One of the main directions of advancement of integrated and fiber optics is the development and production of a wide class of optical elements on a surface and in the interior volume of transparent materials (glass) for converting and controlling optical signals. Such optical elements are obtained by local changes in the optical properties of a material, i.e., by producing regions with modified optical properties. New methods of forming modified regions (MRs)  $1-300 \, \mu m$  in size in the interior volume of materials transparent to the forming radiation have been developed rapidly in the last few years [1, 2]. Laser technologies make it possible to produce MRs with prescribed sizes and shapes by controlling the energy characteristics of the radiation used to form the MRs and the results obtained are highly reproducible, which is accomplished by means of feedback on the optical characteristics in the course of the formation of an MR. All MRs formed on a surface and in the interior volume of glass plates can be conventionally grouped as follows:

- 1) with constant index of refraction different from that of the main material;
  - 2) with an index gradient;
- 3) complexly structured, comprised of parts with different refractive indices that can differ from the index of the main material.

At present special attention is being devoted to the production of complexly structured MRs. A complex structure of an MR permits wider control of the optical properties of the MR and makes it possible to produce optical elements with ultrashort focal lengths.

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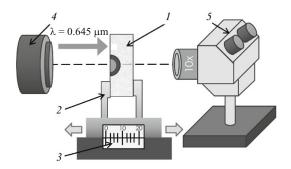
It has been reported in the last few years that lasers with ultrashort pulses  $(10^{-9}-10^{-14}\,\text{sec})$ , high repetition frequency  $(100-250\,\text{kHz})$  and long wavelength radiation for which the glass is transparent ( $\lambda=0.8\,\mu\text{m}$ ) are usually used to form MRs in the interior volume of optically transparent materials [3-7]. Because of the high power density  $(10^{10}-10^{15}\,\text{W/cm}^2)$  in the waist of the laser beam a nonlinear change occurs in the absorption, as a result of which the optical properties in the region of the neck change, i.e., MRs with altered optical properties are formed.

However, information on the formation of MR in the interior volume of a plate of porous glass (PG) by laser radiation with wavelength  $\lambda = 0.808 \, \mu m$ , average power  $P = 120 \, mW$  and linear polarization of the beam is presented in [7, 8]. For long-time stabilization of the optical properties plates with an MR were baked at temperature  $870 \pm 10^{\circ} C$  for  $15 \pm 5 \, min$  to convert the plate into quartzoid. It follows from these reports that an MR in plates of quartzoid glass (QG) can be used as a single optical element for focusing radiation or matching fibers in optoelectronics. We note that in terms of the physical-chemical properties quartzoid glass is only slightly inferior to quartz glass [9]. The quartzoid glass is transparent from the near- to mid-IR ranges and possesses high chemical, thermal and radiation resistance.

It is known that the geometric dimensions and index of refraction of MRs and the refractive index of the plate change when the plate is baked [8, 10, 11], but baking preserves an MR in a PG plate and increases the temporal stability of the optical properties of an optical element. In [8, 11] the optical properties of complexly structure MRs have never been determined for a PG plate or a QG plate obtained by baking PG plates with an MR in a furnace. At the same time

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**Fig. 1.** The experimental arrangement for measuring the focal segment: l) QG plate with an MR; 2) coordinate table; 3) microscope scale; 4) laser module; 5) microscope.

it is precisely the optical properties that determine the potential possibilities of using MRs in different spheres of microsystems engineering. The determination of the optical properties of MRs in PG plates will make it possible to determine the changes occurring in these properties as result of baking and will shed light on how laser radiation weakly absorbed by the plate material forms an MR as well as on the processes occurring in complexly structured MRs when plates with an MR are baked in a furnace.

The objective of the present work is to determine the optical properties of a complexly structured MR in a PG plate and the changes occurring in these properties when a PG plate with an MR is baked in a furnace.

The main optical properties of an MR intended for focusing radiation are: the focal length f, the numerical aperture (NA) or the resolving power N. The optical properties of any optical element, including an MR, are determined by its shape, structure, refractive index and index gradient, if one is present.

The focal length of an MR in a PG plate was determined by measuring the focal segments of thin micro-optic elements, where by using an optical microscope a focal segment could be taken as the focal length [13].

We used this method because in the practice of optical measurements other methods recommended for measuring the focal length, even for optical elements several millimeters in size, are not applicable for objects smaller than 200  $\mu$ m located in the interior volume of a plate at a depth of the order of 400  $\mu$ m.

The method of measuring a focal segment under a microscope is based on measuring the distance from the surface of an MR to the plane with the smallest cross section of a laser beam with small divergence of the radiation passing through the MR. The plane with the smallest laser beam cross section was identified with the focal plane of the MR. By bringing the best-view plane of the microscope into coincidence with the focal plane of the MR being tested it was also possible to determine the size  $d_f$  of its focal spot as well as the quality of the MR, to a first approximation, according to the shape of the focal spot.

The experimental arrangement for measuring the focal segment  $S_f$  and the focal spot  $d_f$  is presented in Fig. 1.

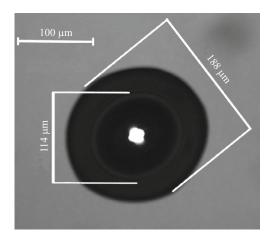
In this method the focal segment was measured as follows. The QG plate 1 with an MR was placed on the coordinate table 2 with a microscope scale 3, where movement along the optic axis of the MR with accuracy ±1 µm was possible. When the QG plate with an MR was positioned its optic axis was in coincidence with the optical axis of the laser module 4 with radiation wavelength  $\lambda = 0.645 \,\mu m$  and the optical axis of the microscope 5 with magnification  $\times 100$ . The surface of the MR was brought into coincidence with the best-view plane of the microscope by moving the coordinate table 2. This position was fixed on the scale 3, along which the coordinate table moved. Next, the coordinate table 2 was moved until the smallest light spot was no longer visible in the best-view plane of the microscope. The displacement of the table between the fixed positions was identified with the focal segment of a thin MR, for which the focal segment  $S_f$ differed very little from the focal length. The accuracy of this method is relatively low, but the method is simply executed and makes it possible to determine the two most important optical characteristics of the MR — the focal length f and the size  $d_f$  of the focal spot.

It follows from the description of the method that the measurement error for f of a spherical MR will be considerable larger than for a hemispherical MR, especially for an MR in the form of a plano-convex lens.

The focal length of a spherical MR in a PG plate was measured 10 times on a setup assembled according to the scheme shown in Fig. 1. For a spherical MR the first reading on the scale was fixed when the central section of the sphere was coincident with the microscope's best-view plane. The measurement error (confidence interval with confidence probability 0.95) did not exceed 7%. The focal length determined from the measurements was  $f = 260 \pm 20 \, \mu m$ .

Since the main problem here was still the determination of the optical properties of an MR in a QG plate, because it is precisely these properties that make it possible to determine the potential application of MRs, the PG plates were not ground to a thickness at which the MR thickness is approximately equal to or less than the radius of a spherical MR. Grinding was also not done because the operation of grinding a PG can give rise to significant mechanical stresses which cause the plates to fracture. For plates exposed to laser radiation the grinding operation is doubly dangerous and usually ends with the MR being spiked out of the plate [9].

The optical properties of a complexly structured MRs in the interior volume of a PG plate were determined for a known geometry and refractive indices of its parts. The sizes of the MR parts were determined from a photograph of a prescribed section of the MR. The photographs were obtained with a Carl Zeiss Axio Imager A1 optical microscope in transmitted light with magnification ×100 (Fig. 2); the MR diameter  $d_{\rm MR}=185~\mu{\rm m}$  and central part diameter  $d_{\rm c,p}=114~\mu{\rm m}$ . The porosity of the MR parts is determined in [11].



**Fig. 2.** Photograph of an MR formed in the interior volume of a PG plate, in transmitted light.

The refractive indices of the MR parts were calculated from the relation  $n_{\rm c,p}=1.414$  and the relation  $n_{\rm s,l}=1.316$  for its surrounding spherical layer; the index for the PG plate is  $n_{\rm pl}=1.342\pm0.005$ . The computed focal length of the MR was f=260.24 µm.

Comparing the computed and measured focal lengths of an MR in a PG plate showed that the discrepancies fall within the measurement error.

The diameter  $d_f$  was determined to be  $d_f$ = 12 ± 0.5 µm according to the photograph of the smallest cross section of the laser beam passing through an MR in the plane (Fig. 3) which we identified with the focal plane of the MR. The numerical aperture of the MR in the PG plate with  $d_{\rm MR}$  = 185 µm and f = 260 µm was 0.36.

To determine the optical properties of a complexly structured MR in a QG plate it is necessary to know its parts and the refractive indexes of these parts. If the sizes of the MR parts under a microscope can be measured quite easily, determination the refractive index of the parts of an MR with diameter about  $160 \mu m$ , located in the interior volume of the glass at depth  $400 \mu m$ , is a difficult technical problem.

It is impossible to determine the refractive index of objects as small as the central region of an MR ( $d_{\rm c,p}$  =  $50-60~\mu m$ ) and the enclosing spherical layer of width  $d_{\rm c,l}$  =  $55-60~\mu m$  by refractometric measurements of n [14], which are widely used in optical measurements. The existing methods of determining the refractive index, which are based on, for example, measurement of the reflection coefficient at a prescribed wavelength, as a rule,  $\lambda = 0.633~\mu m$ , which is the wavelength of single-mode He–Ne laser radiation incident in a direction perpendicular to the plane-parallel surface of the experimental object, are inapplicable for objects of such small size and shape.

However, the refractive indices of parts of an MR can be found. To do this we turn to the results obtained in [11], where porosity data are presented for PG plates  $\delta_{PG}=0.25~cm^3/cm^3$ , the central part of an MR  $\delta_{c,p}=0.102~cm^3/cm^3$  and the

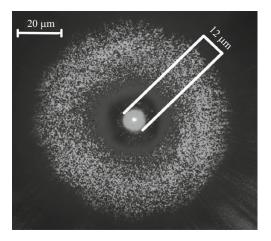


Fig. 3. Photograph of the focal plane of an MR in PG.

spherical layer enclosing the central part  $\delta_{\rm s,l}=0.297~{\rm cm^3/cm^3}$  at a time before the PG with the MR is baked in a furnace. It is also shown in [11] that the porosity of the central part corresponds to its pore volume  $V_{\rm pore~c,p}=0.0235V_{\rm sp}$ , where  $V_{\rm sp}$  is the MR volume, while the porosity  $V_{\rm pore~sp.l}$  of the spherical layer is found from the relation

$$V_{\text{pore sp.l}} = 0.2275 V_{\text{sp}}.$$
 (1)

Over the same period of time that a PG plate compacting to quartzoid shrinks to 0.25 of the initial volume of the plate the MR as a whole also will shrink to 0.25 of the initial volume. The full shrinkage, i.e., compaction of the central part of the MR to quartzoid, having a much lower porosity than that of the PG plate, will definitely occur. Over the compaction time of the PG plate and central part of the MO, for which their porosity and pore volume become vanishing small, a pore volume will remain in the spherical layer; this pore volume can be found as the difference of the pore volume of the spherical layer and the pore volume of the spherical part of the MR at the moment before compaction, i.e.,

$$V_{\text{pore sp.comp}} = V_{\text{pore c.l}} - V_{\text{pore c.p}} = 0.204 V_{\text{sp}}, \qquad (2)$$

where  $V_{\rm pore\ sp.comp}$  is the pore volume in the spherical layer after compaction of the PG plate with MR in a furnace.

The dependence of the pore volume in the spherical layer, expressed in parts of the MR volume, on the volume of the spherical layer, also expressed in parts of the MR volume, i.e.,  $V_{\rm pore\; sp.l.}=f(V_{\rm sp.l})$ , is presented in Fig. 4. Given the pore volume in the spherical layer, the known volume of the spherical layer, the known volume of the spherical layer at the start of the compaction process and the known volume of the spherical layer at the end of the compaction process (see Fig. 4) the volume of the spherical layer  $V_{\rm sp.comp}$  after compaction can be determined from the volume of the pores remaining in the spherical layer with complete shrinkage of the central part of the MR and the plate as a whole on the basis of the expression

$$V_{\text{sp.comp}} = 0.742 V_{\text{sp.}} \tag{3}$$

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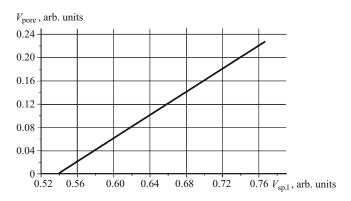


Fig. 4. Pore volume  $V_{\text{pore}}$  of a spherical layer versus the volume  $V_{\text{sp.1}}$ .

Given the volume of the pores remaining in the spherical layer and the volume of the spherical layer the porosity corresponding to a given pore volume  $\delta_{sp.comp}$  can be calculated from the expression

$$\delta_{\text{sp.comp}} = \frac{V_{\text{poresp.comp}}}{V_{\text{sp.comp}}}.$$
 (4)

The value found  $\delta_{\rm sp.comp} = 0.275~{\rm cm}^3/{\rm cm}^3$  made it possible to determine the refractive index of the spherical layer of the MR in a QG plate  $n_2 = 1.331$  from the relation  $n = f(\delta)$ , presented in [12] for PG plates, whose composition and processing were close to that of PG plates used in the experiment on the formation of MRs.

Since before the measurements were performed the QG plate was ground to a thickness at which the MR thickness was approximately equal to the radius of a spherical MR in order to decrease the error of measurement for the focal segment, the optical characteristics of the complexly structured MR in a QG plate were determined according to the known geometry of the parts of a hemisphere and according to the known refractive indices of these parts.

The QG plates were ground and polished using Buehler MetaServ 250 equipment. Since baking reduces thermo-

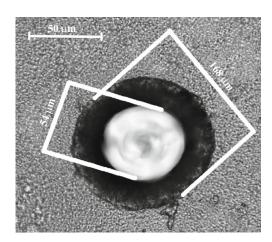
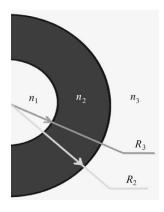


Fig. 5. Photograph of a polished MR in QG in transmitted light.



**Fig. 6.** Schematic image of MR in QG.

mechanical stresses present in PG plates exposed to laser radiation [9], the mechanical processing was successful, no cracking or spike out of MR occurred.

The sizes of the MR parts were determined by photographing the MR cross sections (Fig. 5), which was accomplished in transmitted light in a Carl Zeiss Axio Imager A1 microscope with ×100 magnification; the values were found to be  $d_{\rm MR} = 168~\mu {\rm m}$  and  $d_{\rm c.p} = 54~\mu {\rm m}$ . The refractive indices of the QG plate  $n_3$  and the central part of the MR  $n_1$ , which also shrunk completely, are equal to 1, and their values for wavelength  $\lambda = 0.633~\mu {\rm m}$  can be taken as  $n_1 = n_2 = 1.458$ . The refractive index  $n_2$  of the spherical layer, enclosing the central part of the MR, was found to be  $n_2 = 1.331$  (Fig. 6).

The hemispherical MR, whose flat surface is coincident with the surface of a polished QG plate, can be represented as an optical system consisting of three thin optical components [15]. In accordance with the rules for signs adopted in geometric optics the focal length of such an MR can be found from the expression

$$f = \frac{n_2 R_2 R_3}{(R_3 - R_2)[n_2(n_1 - 2) + 1]},$$
 (5)

where  $R_2$  is the radius of the MR and  $R_3$  is the radius of its central part after the PG plate with an MR is baked in a furnace.

For  $R_2 = 84 \,\mu\text{m}$  and  $R_3 = 27 \,\mu\text{m}$  the focal length of a hemispherical MR is  $f = 188 \,\mu\text{m}$ .

The focal length of the MR in a QG plate was measured by the method of measuring a focal segment under a microscope. The focal length of a hemispherical MR in a QG plate was measured 10 times in a model of the setup. The measurement error for f (the confidence interval with confidence probability 0.95) did not exceed 5%. The focal length determined as a result of the measurements was  $f = 190 \pm 10$   $\mu$ m. The value  $d_f = 7.0 \pm 0.5$   $\mu$ m was obtained from a photograph of the smallest cross section of the laser beam passing through the MR, which we identified with the focal length of the MR, taking account of the scale of the photograph (Fig. 7).

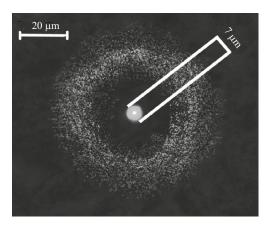


Fig. 7. Photograph of the focal plane of the MR in QG.

The results of the experimental measurements and calculation of the focal length were in agreement with one another to within the measurement error.

Now, when the sizes of the MR parts and the refractive indices of these parts and the QG plate are known, the focal length  $f_{\rm sp}$  of a spherical MR can be calculated using well-known relations from geometric optics (5) assuming that both hemispherical components  $f_{\rm h.sp}$  of the MR can be regarded as thin and therefore  $f_{\rm sp} = f_{\rm h.sp}$ .

The numerical aperture was determined, according to the known focal length  $f_{\rm sp}=90~\mu{\rm m}$  and diameter of the MR, to be NA = 0.9. We did not expect such a high value of the NA, since the stabilization of the properties of thermally compacted optical elements produced on the surface of PG plates by CO<sub>2</sub> laser radiation during baking in a furnace was always accompanied by a decrease of the numerical aperture. The optical elements of the MR in a QG plate were found to be much more attractive from the standpoint of using MR than the analogous properties of MR in a PG plate.

The significant improvement of all optical properties of MRs, first and foremost, the numerical aperture by a factor of 2.5, which is the most important property of an MR, determining its functional possibilities, in the process of baking PG plates with MR in a furnace gives hope that MRs will find the widest possible application in unprecedentedly rapidly growing areas of fiber-optic communication, integrated optics and microsystems engineering.

## **CONCLUSIONS**

The optical properties of complexly structured MRs in the interior volume of PG and QG plates were determined. The focal length of MRs in both cases was determined experimentally and computationally. In both cases, to calculate the focal length of MRs in a QG plate the refractive index of the spherical layer enclosing the central part of the MR was determined using the pre-baking porosity for the MR parts. The focal length of an MR in a QG plate was measured on QG plates polished to thickness approximately equal to the radius of the MR in order to decrease the measurement error.

The computational and experimental results in both cases were in agreement with one another to within the measurement error. The MR in the QG plate shows quite high resolution > 100 lines/mm and an unexpectedly large numerical aperture NA = 0.9. As far as we know, up till now there have been no reports of such large numerical apertures having been obtained in a single optical element.

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## REFERENCES

- R. Jason, A. Luis, and Peter R. Herman, "Femtosecond laser writing of optical edge filters in fused silica optical waveguides," *Optics Express*, 21(4), 4493 – 4502 (2013).
- Yu Teng, Jiajia Zhou, and Geng Lin, "Ultrafast modification of elements distribution and local luminescence properties in glass," J. Non-Cryst. Solids, 358, 1185 – 1189 (2012).
- 3. M. C. Estevez, M. Alvarez, and L. M. Lechuga, "Integrated optical devices for lab-on-a-chip biosensing applications," *Laser & Photonics Rev.*, **6**(4), 463 487 (2012).
- 4. R. Osellame, G. Cerullo, and R. Ramponi, Femtosecond Laser Micromachining: Photonic and Microfluidic Devices in Transparent Material, Springer, Berlin (2012).
- G. Della Valle, R. Osellame, and P. Laporta, "Micromachining of photonic devices by femtosecond laser pulses," *J. Opt. A: Pure Appl. Opt.*, No. 11, 013001 (2009).
- Yang Liao, Jiangxin Song, and En Li, "Rapid prototyping of three-dimensional microfluidic mixers in glass by femtosecond laser direct writing," *Lab. Chip.*, 12, 746 – 749 (2012).
- 7. G. K. Kostyuk, M. M. Sergeev, T. V. Antropova, et al., "Laser-induced local change in the optical properties of borosilicate glasses," *Fiz. Khim. Stekla*, No. 3 (2013).
- G. K. Kostyuk, M. M. Sergeev, T. V. Antropova, et al., "Laser induced structural changes in porous glass by hot and cold compaction," *Steklo Keram.*, No. 12, 3 7 (2012); G. K. Kostyuk, M. M. Sergeev, T. V. Antropova, et al., "Laser induced structural changes in porous glass due to hot and cold compaction," *Glass Ceram.*, 69(11 12), 393 396 (2012).
- 9. O. V. Mazurin, G. P. Roskova, V. I. Aver'yanov, and T. V. Antropova, *Biphase Glasses: Structure, Properties and Applications* [in Russian], Nauka, Leningrad (1991).
- T. V. Antropova, V. P. Veiko, and G. K. Kostyuk, "Particulars of the formation of planar micro-optical elements on porous glass substrates by laser radiation followed by sintering," *Fiz. Khim. Stekla*, 38(6), 699 – 717 (2012).
- G. K. Kostyuk, M. M. Sergeev, and E. B. Yakovlev, "Nature of modified regions produced in the interior volume of glass by laser radiation with wavelength weakly absorbed by the glass," *Perspekt. Mater.*, No. 9, 43 – 53 (2013).
- 12. G. K. Kostyuk, V. P. Veiko, G. P. Roskova, et al., "Refractive indices of high-silica porous glasses with different porosity," *Fiz. Khim. Stekla*, **15**(2), 213 238 (1989).
- V. P. Veiko, G. K. Kostyuk, G. P. Roskova, and T. S. Tsekhomskaya, *Laser Formation of Microoptic Elements* [in Russian], LDNTP, Leningrad (1988).
- B. V. Ioffe, Refractometric Methods in Chemistry [in Russian], Leningrad (1974), 2nd edition.
- B. N. Begunov, N. P. Zakaznov, S. I. Kiryushin, and V. I. Kuzichev, *Theory of Optical Systems* [in Russian], Mashinostroenie, Moscow (1981).